UCRL-JC-126889 Abs

Abstract for Actinide Separations Conference

Plutonium Immobilization: Ceramic Process Development & Demonstration

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This paper describes process development activities for the Fissile Materials Immobilization Program (FMIP). Zirconolite and pyrochlore forumlations developed by Van Konynenburg and Ebbinghaus are being used [1997]. The stoichiometric formula of the zirconolite material is $(Ca_{0.88}Gd_{0.12})(Zr_{0.88}Pu_{0.12})(Ti_{1.78}Al_{0.22})O_7$, with 5 wt. % excess rutile and 5 wt. % barrium hollandite. The pyrochlore formula is $(Ca_{0.89}Gd_{0.11})(Hf_{0.23}U_{0.44}Pu_{0.22}Gd_{0.11})Ti_2O_7$ and has 5 wt. % excess rutile, as well as 1 wt. % excess hafnia. Ceria (CeO_2) has been used as a surrogate for PuO_2 in all cold (non-radioactive) process development. Unit operations required for the synthesis of these materials include: (1) drying precursors; (2) calcining precursors; (3) wet mixing and drying of precursors; (4) jet milling oxide, $PuO_2/UO_2/CeO_2$; (4) V-blending and granulation of pre-mixed precursors, $PuO_2/UO_2/CeO_2$, and PEG-water binder; (5) cold pressing of granulated material; (6) reactive sintering; and (7) measurement, testing, and evaluation. Typically, drying is done in air at 90 °C, while calcining is done in argon at 750°C for 1 hour. The precursors are then wet mixed for 1 hour. Oxide ($PuO_2/UO_2/CeO_2$) feed is milled to the desired particle sizes by a small, efficient jet mill. Size reduction results from particle collisions in two impinging jets; one particle is attrited by another. This device was modified at LLNL to include two cyclone separators and a cylindrical HEPA filter connected in series. The cummulative separation efficiency of the two cyclones is approximately 95 percent. Particles not removed by the cyclones are removed by the HEPA; no particles are detected downstream of the HEPA. Tests with -150 to +325 mesh TaO_2 showed that a product with an average particle size of 0.3 to 0.5 μm could be produced at a throughput of 1 kg hr^{-1}. Similar results have now been obtained with PuO_2. The wet-mixed precursors, milled oxide ($PuO_2/UO_2/CeO_2$), and PEG-water binder are then V-blended and granulated. Granules are cold pressed in an automated hydraulic press with a 2-punch die at approximately 2800 to 3000 psi. The entire pressing cycle is only 2 to 3 minutes in duration. Sintering, the final step, is done in argon at 1300 to 1350 °C for 4 hours. Ceramic forms with measured densities as great as 97 to 98% theoretical have been produced. Experience from the MOX industry is being used to enhance process design.

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.